



08/30/00

US 35 U.S.

PTO

UTILITY PATENT APPLICATION TRANSMITTAL

(Only for new nonprovisional applications under 37 CFR 1.53(b))

Attorney Docket No.

684.3060

First Named Inventor or Application Identifier

Yukio Hanyu

Express Mail Label No.

APPLICATION ELEMENTS

See MPEP chapter 600 concerning utility patent application contents.

ADDRESS TO:

Assistant Commissioner for Patents
Box Patent Application
Washington, DC 20231

1. ☐ Fee Transmittal Form
(Submit an original, and a duplicate for fee processing)
2. ☒ Specification Total Pages
3. ☒ Drawing(s) (35 USC 113) Total Sheets
4. ☐ Oath or Declaration Total Pages
- a. ☐ Newly executed (original or copy)
- b. ☐ Unexecuted for information purposes
- c. ☐ Copy from a prior application (37 CFR 1.63(d))
(for continuation/divisional with Box 17 completed)
[Note Box 5 below]
- i. ☐ DELETION OF INVENTOR(S)
Signed Statement attached deleting
inventor(s) named in the prior application, see
37 CFR 1.63(d)(2) and 1.33(b).
5. ☐ Incorporation By Reference (useable if Box 4c is checked)
The entire disclosure of the prior application, from which a copy of
the oath or declaration is supplied under Box 4c, is considered as
being part of the disclosure of the accompanying application and is
hereby incorporated by reference therein.

6. ☐ Microfiche Computer Program (Appendix)
7. Nucleotide and/or Amino Acid Sequence Submission
(if applicable, all necessary)
- a. ☐ Computer Readable Copy
- b. ☐ Paper Copy (identical to computer copy)
- c. ☐ Statement verifying identity of above copies

ACCOMPANYING APPLICATION PARTS

8. ☐ Assignment Papers (cover sheet & document(s))
9. ☐ 37 CFR 3.73(b) Statement ☐ Power of Attorney
(when there is an assignee)
10. ☐ English Translation Document (if applicable)
11. ☐ Information Disclosure Statement (IDS)/PTO-1449 ☐ Copies of IDS Citations
12. ☐ Preliminary Amendment
13. ☒ Return Receipt Postcard (MPEP 503)
(Should be specifically itemized)
14. ☐ Small Entity Statement(s) ☐ Statement filed in prior application
Status still proper and desired
15. ☐ Certified Copy of Priority Document(s)
(if foreign priority is claimed)
16. ☒ Other: Inventor Information Sheet

17. If a CONTINUING APPLICATION, check appropriate box and supply the requisite information:

☐ Continuation ☐ Divisional ☐ Continuation-in-part (CIP) of prior application No. /

18. CORRESPONDENCE ADDRESS

☒ Customer Number or Bar Code Label

05514

(Insert Customer No. or Attach bar code label here)

or ☐ Correspondence address below

NAME

Address

City

State

Zip Code

Country

Telephone

Fax



CLAIMS	(1) FOR	(2) NUMBER FILED	(3) NUMBER EXTRA	(4) RATE	(5) CALCULATIONS
	TOTAL CLAIMS (37 CFR 1.16(c))	11 -20 =	0	X \$ 18.00 =	\$0.00
	INDEPENDENT CLAIMS (37 cfr 1.16(b))	2 - 3 =	0	X \$ 78.00 =	\$0.00
	MULTIPLE DEPENDENT CLAIMS (if applicable) (37 CFR 1.16(d))			\$260.00 =	\$260.00
				BASIC FEE (37 CFR 1.16(a))	\$690.00
				Total of above Calculations =	\$950.00
	Reduction by 50% for filing by small entity (Note 37 CFR 1.9, 1.27, 1.28).				
	TOTAL =				\$950.00

19 Small entity status

- a. ☐ A Small entity statement is enclosed
- b. ☐ A small entity statement was filed in the prior nonprovisional application and such status is still proper and desired.
- c. ☐ Is no longer claimed.

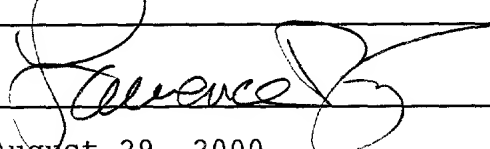
20 ☒ A check in the amount of \$ 950.00 to cover the filing fee is enclosed

21 ☐ A check in the amount of \$ _____ to cover the recordal fee is enclosed.

22. The Commissioner is hereby authorized to credit overpayments or charge the following fees to Deposit Account No. 06-1205:

- a. ☒ Fees required under 37 CFR 1.16.
- b. ☒ Fees required under 37 CFR 1.17.
- c. ☐ Fees required under 37 CFR 1.18.

SIGNATURE OF APPLICANT, ATTORNEY, OR AGENT REQUIRED

NAME	Lawrence S. Perry Registration No. 31,865
SIGNATURE	
DATE	August 29, 2000

[illegible]

SINGLE-CRYSTALLINE FILM
AND PROCESS FOR PRODUCTION THEREOF

FIELD OF THE INVENTION AND RELATED ART

5 The present invention relates to a molecular
single-crystalline film (which herein refers to a film
having a thickness of at most ca. 100 μm and having a
portion which retains a single crystal state having a
uniform molecular crystalline alignment over the
10 thickness and over an areal extension including a side
length of at least 10 times the thickness, i.e., an
areal size utilizable as a functional film, preferably
an areal size of at least 50 μm x 50 μm), and a
process for production thereof.

15 A molecular crystal can be expected as a
useful device material, such as a superconducting
material, an effective photoconductor or a gas sensor,
because of its electronical and geometrical structure
and packing state. As the process for production
20 thereof, growth in a solution and growth in a molten
state have been generally practiced. According to any
of such processes, however, it is difficult to obtain
a thin film of single crystal by suppressing the
thickness increase, and this poses an obstacle against
25 using it as a functional layer in devices which have a
laminar structure in many cases. As another process,
there is known a gas phase deposition process, by

which, however, it is difficult to prepare a uniform film due to affection by the gas phase deposition boundary.

On the other hand, it has been reported to
5 improve the carrier transportation performance by
utilizing a molecular alignment in a higher-order
liquid crystal phase of SmB or SmE ("Ohyou Butsuri
(Applied Physics)", Vol. 68, No. 1, pp. 26 - 32
(1999)). In this report, a higher speed
10 transportation of electrons and holes has been aimed
at by utilization of alignment order in a higher-order
liquid crystal phase. The improvement in high-speed
transportation performance has been considered
attributable to the formation of flow paths for
15 electrons and holes due to regular packing of aromatic
rings in the higher-order smectic phase alignment.
This performance has been also noted as a carrier
transportation layer in EL devices, and a further
improvement is expected.

20 Regardless of whether it is a liquid crystal
or a (solid) crystal (herein a term "crystal" without
further notation is used to mean a solid crystal), the
film thereof is required to assume a single crystal
state free from defects (i.e., free from carrier
25 traps) in order to function as a functional layer as
mentioned above.

Then, if a (solid) single-crystalline film

can be obtained, it is expected to achieve a higher-speed and higher-density carrier transportation because of a higher degree of order and a closer packaging of molecules than a liquid crystal film.

5

SUMMARY OF THE INVENTION

In view of the above-mentioned circumstances, a principal object of the present invention is to provide a molecular single-crystalline film usable in a device, and a process for effective production thereof.

In order to achieve the above-mentioned object, it may be conceived to form a liquid crystal material layer of which the thickness is regulated between a pair of boundaries at a higher temperature and cool the liquid crystal material layer to room temperature, thereby forming a crystal layer wherein the molecular alignment is fixed. As a result of my study, however, such a crystal film obtained through the above-described process in general can only form a poly-crystalline film and fails to provide a single-crystalline film. This is considered because a strain or disclination in a domain relaxed in a liquid crystal phase because of fluidity or flexibility of the liquid crystal phase is developed to cause precipitation of crystallites or polycrystallization during the crystallization.

5 boundaries can be phase-transformed into a single-crystalline film while remarkably suppressing the polycrystallization.

I have also discovered a smectic liquid crystal material exhibiting a uniform (i.e., a single mode of) molecular alignment inclusive of a director (i.e., molecular long-axis) direction in a smectic layer as a suitable material as the above-mentioned liquid crystal material having a better regularity.

a step of disposing a smectic liquid crystal material exhibiting a uniform molecular alignment in a smectic layer between a pair of boundaries having a thickness regulation function, and

a crystallization step of cooling and

apparent upon a consideration of the following description of the preferred embodiments of the present invention taken in conjunction with the accompanying drawings.

5

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 is a schematic thicknesswise sectional view illustrating a basic structure of a cell used for formation therein of a single-crystalline film according to the invention.

10

Figure 2 illustrates a deviation in alignment of aromatic rings in a random stacking of head-tail molecules.

Figure 3 illustrates a better overlapping of aromatic rings in a stacking of head-head molecules.

15

Figure 4 is a polarization microscope photograph (x100) of nematic phase (130 °C) in a cell of Example 1.

Figure 5 is a polarization microscope photograph of smectic C phase (123 °C) in the cell of Example 1.

20

Figure 6 is a polarization microscope photograph of crystal phase (30 °C) in the cell of Example 1.

25

Figures 7 and 8 are polarization microscope photographs of states after 1 minute and 10 minutes, respectively, held at 118 °C in a cell of Example 2.

Figure 9 is a schematic view based on a sketch of the photograph of Figure 6.

Figure 10 illustrates an outline of an X-ray diffraction apparatus for examining the crystallinity of a film in a sample cell.

Figure 11 is a graph showing X-ray diffraction patterns (at an X-ray incidence angle $\alpha = 95$ deg.) of a single crystal portion (s-crystal) and a polycrystal portion (p-crystal), respectively, in a cell of Example 1A.

Figures 12 and 13 are graphs showing changes of X-ray diffraction patterns with variation of incidence angles ($\alpha = 80 - 110$ deg.) of the single crystal portion (s-crystal) and polycrystal portion (p-crystal), respectively, in the cell of Example 1A.

Figure 14 is a polarizing microscope photograph (x150) showing the crystallinity of a film in a cell of Example 2A.

Figure 15 is a polarizing microscope photograph (x75) showing the crystallinity of a film in a cell of Example 5.

DETAILED DESCRIPTION OF THE INVENTION

According to an embodiment, a single-crystalline film according to the present invention may be prepared in a structure of cell (device) as illustrated in a schematic sectional view of Figure 1

which is at a glance similar to that of a conventional liquid crystal cell.

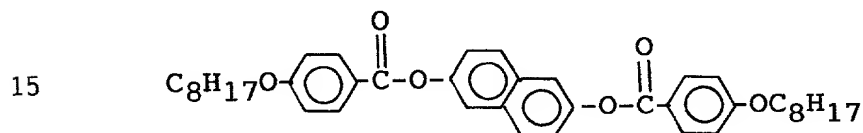
Referring to Figure 1, the cell structure includes a pair of glass substrates 1a and 1b having thereon transparent electrodes 22a and 22b, respectively, of ITO (indium tin oxide), etc., and alignment control films 23a and 23b, respectively, of 50 to 1000 Å-thick polyimide film, etc., disposed opposite to each other with a gap therebetween determined by a spacer 12 disposed therebetween, and a single-crystalline film 13 formed between the substrates. More specifically, for the preparation, a blank cell structure excluding the single-crystalline film 13 may be prepared first similarly as in the preparation of an ordinary liquid crystal cell, a liquid crystal material showing fluidity by heating may be injected into the cell to seal up a liquid crystal layer 13 in the cell, and the liquid crystal layer 13 may be gradually cooled to form a single-crystalline film 13.

As described above, the liquid crystal material constituting the film 13 is required to have a liquid crystal phase having a good regularity. An example thereof is a smectic liquid crystal material providing a uniform molecular alignment in a smectic layer, and a suitable example is a smectic liquid crystal material having a molecular structure which is

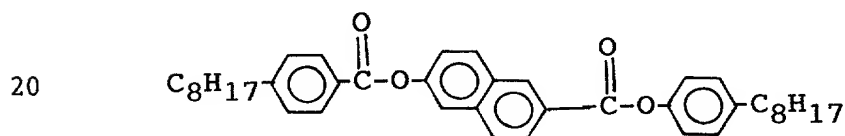
symmetrical with respect to the molecular long axis.
Specific examples thereof may include smectic liquid
crystal materials including a material used in
Examples described hereinafter and represented by the
5 following general formula (1):



wherein M1 denotes a laterally symmetrical mesogen
(i.e., mesomorphic core) unit, and R1 denotes a
terminal chain group, such as an alkyl or an alkoxy
10 group suitable for providing a smectic liquid crystal
phase. Specific examples of smectic liquid crystal
materials represented by the formula (1) may include
the following compounds:



Iso $\xrightarrow{234.4}$ N $\xrightarrow{141}$ SmC $\xrightarrow{107}$ Crystal



Iso $\xrightarrow{213.7}$ SmA $\xrightarrow{133.6}$ (SmC) \longrightarrow Crystal

As described above, an asymmetrical smectic
25 liquid crystal material as represented by a formula
(2) below:



wherein M2 denotes a mesogen unit, and R2 and R3 denote mutually different terminal chain groups suitable for providing a smectic liquid crystal phase, is not generally suitable for the object of the present invention. However, it is possible to use a kind of smectic liquid crystal material, such as one characterized by, e.g., a combination wherein R2 is an alkyl(oxy) chain, and R3 is a group which is repulsive from R2 and per se shows a strong aggregatability, such as a fluoroalkyl(oxy) chain, in the present invention, because such a smectic liquid crystal material can form a stack of uniformly aligned molecules wherein the groups R2 are directed along one side and the groups R3 are directed along the other side of a smectic layer, thus consequently providing a stack of uniformly aligned molecules R2-M2-R3 in a smectic layer and being less liable to form crystal defects at the time of crystallization.

A liquid crystal material having a uniform molecular alignment in a smectic layer advantageously affects the properties of the resultant single-crystalline film. More specifically, in the case of an alignment wherein a head-tail molecule and a tail-head molecule are stacked at random, the resultant single-crystalline film even if formed as such is caused to include a shift of aromatic rings constituting the liquid crystal material as shown in

Figure 2. In contrast thereto, in a single-crystalline fine formed by stacking of molecules having a symmetrical structure, such as a head-head, it becomes possible to obtain better electrical and optical properties attributable to overlapping of π -electrons as illustrated in Figure 3.

As the single-crystalline film of the present invention is provided with a molecular alignment order through phase transition from a liquid crystal phase, it is preferred that the liquid crystal material used in the present invention has at least one liquid crystal phase at a higher temperature region than room temperature and is cooled to provide a stable crystal film at room temperature. It is further preferred that the liquid crystal material used has two or more liquid crystal phases and is caused to enhance the alignment order in the course of cooling from a lower order of liquid crystal phase to a higher order of liquid crystal phase and be crystallized into a single-crystalline film as a result of further higher degree of order. A preferred example of phase transition series to be assumed by the liquid crystal material may include the following:

Cryst - SmC - N - Iso.

The single-crystalline film 13 may have a thickness which can be arbitrarily set within a range of, e.g., 100 nm - 100 μ m, preferably ca. 1 - 10 μ m,

depending on the function of the film in the device including the film.

The cooling speed for formation of the single-crystalline film may preferably be at most 10
5 °C/min., more preferably at most 5 °C/min, particularly preferably ca. 1 - 3 °C/min., while it can depend on the thickness of the film formed.

By selection of an appropriate liquid crystal material, the single-crystalline film 13 can be formed
10 through a single course of cooling from such a liquid crystal phase (as shown in Example 1 described hereinafter). However, in order to obtain a single-crystalline film having a better crystallinity and/or including a broader area of single crystal region, it
15 is also preferred to include an operation of reheating a once-formed single-crystalline film again to a crystal region temperature which is close to the liquid crystal - crystal transition temperature, preferably in a range of the transition temperature -
20 10 °C, more preferably in a range of the transition temperature - 3 °C and holding the film at that temperature for a prescribed period of ca. 0.5 - 5 hours. As a result of such an operation, it becomes possible to convert a polycrystalline region remaining
25 in the once-formed single-crystalline film or cause the once-formed single crystal region to grow into a broader region. Incidentally, the holding at a

once cooling to room temperature (as shown in Example 2 described later). In any case, it is possible to obtain a single-crystalline film having a better single crystallinity by cooling to room temperature after the holding.

10 In the embodiment of Figure 1, the thickness
of the film 13 is regulated by the bead spacer 12. It
has been confirmed that the presence of such bead
spacer 12 does not adversely affect the single
crystallinity of the resultant film 13 up to ca. 20
15 μm of the thickness. While it depends on the area of
the film 13, in order to form a thickener film, the
bead spacer can be omitted or replaced by a stripe
spacer.

Incidentally, as will be understood from
20 Examples described later, the transparent electrodes
22a and 22b are unnecessary simply for the purpose of
formation of a single-crystalline film, but the
crystallization can be performed under application of
a voltage as desired. Further, at least in the case
25 of using a smectic liquid crystal material having a
symmetrical molecular structure as represented by the
above-mentioned formula (1) and used in the following

Examples, the presence or absence of alignment films 23a and 23b, or the presence or absence of a uniaxial aligning treatment, such as rubbing, for the alignment films 23a and 23b, does not essentially affect the formation of a single-crystalline film 13.

Accordingly, such alignment films 23a and 23b can be omitted, as desired. Thus, the surfaces of a pair of substrates 1a and 1b contacting the liquid crystal layer 13 injected into the cell can essentially comprise any arbitrary material capable of providing a pair of boundaries for converting the liquid crystal layer 13 into a single crystal while regulating the thickness of the liquid crystal layer 13 at constant.

However, depending on the liquid crystal material used, it is possible to positively utilize the alignment control force of a boundary for aligning liquid crystal molecules perpendicular to, parallel to, or inclined at a desired inclination to the boundary and utilize the alignment order for formation of a single-crystalline film in the crystal phase of a higher degree of order.

As is understood from the above description, a substantial latitude is left regarding the materials constituting a pair of boundaries contacting the liquid crystal layer 13. Accordingly, in the case of using the single-crystalline film 13 in the above embodiment, e.g., as a hole-transporting layer in an

5
10

15
20
25

an arbitrary curvature. Such a substrate having a curved surface may preferably be composed of, e.g., a metal having a good thermal conductivity. As a result, a film device once disposed on a curved
5 surface of substrate may be elevated to an appropriate crystallization temperature, held for a prescribed period at that temperature and then gradually cooled to room temperature, whereby the film in the film device can be converted into a single-crystalline film
10 which per se is in the curved state, thus providing a curved single-crystalline film device.

As is shown in the above-described embodiments, a film formed in situ and incorporated in a cell structure is a preferred embodiment of the
15 single-crystalline film according to the present invention. Depending on a required function thereof, however, such a single-crystalline film according to the present invention formed in situ in a cell structure can be used in a form isolated from such a
20 cell structure or in a form laminated with another functional layer by transferring from such a cell structure.

[Examples]

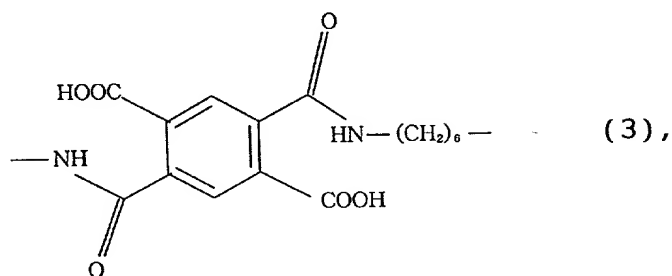
Hereinbelow, the present invention will be
25 described more specifically based on Examples.

(Example 1)

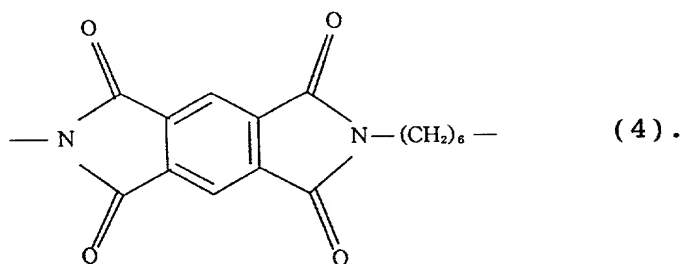
A cell having a layer structure schematically

illustrated in Figure 1 was prepared.

Two glass sheets each having a thickness of 1.1 mm and an areal size of ca. 20 mm x 20 mm were respectively coated with a 700 Å-thick ITO transparent conductor film by sputtering and further with a 0.7 wt. % solution in NMP (N-methylpyrrolidone) of a polyamic acid ("LP-64", made by Toray K.K.) having a recurring unit of formula (3) below by spin coating at 2000 rpm for 20 sec:



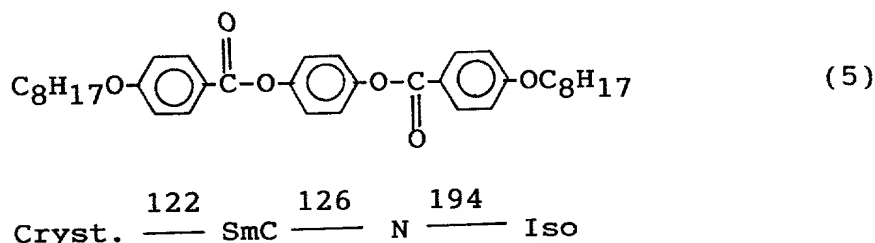
followed by pre-drying at 80 °C for 5 min. and baking at 200 °C for 60 min. to form a 50 Å-thick film of polyimide represented by formula (4) below:



The polyimide film on each glass substrate was subjected to four times of rubbing in one direction with a nylon-planted cloth at a roller feed speed of

10 mm/sec and a roller revolution speed of 1000 rpm.

The two substrates treated in the above-described manner were applied to each other with 2.4 μ m-dia. spacer beads disposed therebetween at a density of 200 beads/mm² to form a blank cell having a cell gap of ca. 2.0 μ m. Then, a liquid crystal material having a structure of formula (5) and a phase transition series respectively shown below was injected into and sealed up within the cell at a nematic phase temperature (130 °C) to form a sample cell, which was then cooled to room temperature at a rate of 1 °C/min., thereby crystallizing the liquid crystal material within cell to form a crystalline film.



In the meantime, the alignment state of the liquid crystal material was observed and photographed through a polarizing microscope in a nematic phase (at 130 °C), a smectic phase (at 123 °C) and in a crystal phase (at 30 °C). The thus-obtained photographs (each at a magnification of 100) are respectively attached hereto as Figure 4 (nematic phase), Figure 5 (smectic phase) and Figure 6 (crystal

phase). Further, a schematic view based on a sketch of the photograph of Figure 6 is attached hereto as Figure 9.

In the photograph of Figure 5, there are
5 observed two-direction domains peculiar to nematic-SmC transition and showing therein microdomains due to slightly different layer directions as an indication of alignment uniformity therein. In the photograph of Figure 6, as indicated in Figure 9 which is a
10 schematic view based on a sketch thereof, a single crystal (s-crystal) region developed to an area of ca. 0.5 mm^2 which is sufficiently large for use as a single crystal functional film is recognized in a left half, while a poly-crystal (p-crystal) region remains
15 in a right half (i.e., crystal state of 50 % uniform alignment), thus showing that a single-crystalline film of the present invention was obtained. Further, it was confirmed by the observation through a polarizing microscope that the single crystal region
20 (in the left of Figure 6) showed an optical uniformity which was remarkably improved than in the SmC alignment (as shown in Figure 5).

(Example 1A)

In order to examine the crystalline order of
25 the respective regions in the crystalline film obtained in Example 1, a cell for X-ray diffraction analysis was prepared in the same manner as in Example

1 except for using 80 μm -thick glass substrates.

The cell was set in a rotary pair cathode-type X-ray diffraction apparatus ("RU-300", made by Rigaku Denki K.K.) having an organization as

5 illustrated in Figure 10 to obtain X-ray diffraction patterns for the single crystal region (s-crystal) and poly-crystal region (p-crystal) respectively in Figure 6 by a transmission method under the following conditions:

10 X-ray source: $\text{CuK}\alpha$, 40 kV, 200 mA

Measurement conditions:

Effective line focus width = 0.05 mm

S1 = 0.15 mm, S2 = SS2 deg., S3 = 0.3 mm,

Ni filter

15 Focus-S1 = 95 mm, S1-Sample = 90 mm,

Sample-S2 = 143 mm, S2-S3 = 42 mm

Incident angle (α deg.) = fixed

2 θ -scan 1 deg./min., Interval = 0.02 deg.

Angle resolution = ca. 3.5 rad (= ca. 0.2 deg.)

20 Sample cell X-ray irradiation region

width = 0.3 mm, length = ca. 10 mm.

X-ray diffraction patterns obtained at a fixed incident angle α = 95 deg. for the single crystal region (s-crystal) and poly-crystal region (p-crystal) are shown in parallel in Figure 11. As shown in Figure 11, two diffraction peaks each were
25 observed at 2θ = 8.5 deg. (d = 10.4 Å) and 2θ = 22.66

deg. ($d = 3.92$ Å) for the single crystal region, and
at $2\theta = 8.8$ deg. ($d = 10.0$ Å) and $2\theta = 22.66$ deg. ($d =$
4.04 Å) for the poly-crystal region.

Then, the X-ray incidence angle was varied at
5 an increment of 5 deg. in the range of 80 - 110 deg.,
and diffraction patterns obtained in the neighborhood
of lower angle side peaks ($\theta = 8.5$ deg. and 8.8 deg.)
are inclusively shown in Figures 12 and 13 for the
single crystal region an the poly-crystal region,
10 respectively. According to Figure 12, the single
crystal region exhibits a maximum peak intensity in
the neighborhood of $\alpha = 95$ deg. and exhibits
substantially no observable peak at $\alpha = 80$ deg. or 110
deg. In contrast thereto, the poly-crystal region
15 shown in Figure 13 generally exhibits a lower peak
intensity than the single crystal region and
substantially no dependence on the incidence angle
change. These results indicate that the single
crystal region exhibits an anisotropy for X-ray
20 diffraction and a higher order of molecular
crystalline alignment, whereas the polycrystal region
exhibits substantially no anisotropy for X-ray
diffraction.

(Example 2)

25 The cell prepared in Example 1 was again
heated to a nematic phase temperature (130 °C) and
thereafter started to be cooled at a rate of 1 °C/min.

similarly as in Example 1. In this Example, however,
the cell was not continually cooled to room
temperature as in Example 1 but held for 30 min. at
118 °C which was a crystal phase temperature lower
5 than the SmC-Crystal transition temperature (= 122 °C)
and thereafter cooled to room temperature. In the
meantime, the alignment states were photographed
through a polarizing microscope after holding for 1
min. and 10 min., respectively, at 118 °C. The thus-
10 obtained photographs (each in a magnification of 100)
are attached hereto as Figures 7 and 8, respectively.

With the lapse of time of the holding at 118
°C, the single crystal region was remarkably enlarged
to ca. 80 % of the cell area (20 mm x 20 mm) as shown
15 in Figure 7 after holding for 1 min., which already
exceeded the single crystal region percentage (ca. 50
%) shown in Figure 6 (obtained by holding for ca. 1.5
hours at crystal region temperatures in Example 1).
Then, as shown in Figure 8, the entire region (100 %)
20 of the cell area was recognized to be single-
crystallized. Incidentally, Figure 8 shows two types
of regions of white and black. It was confirmed that
these two types of regions were respectively single
crystal regions (domains) which had different planar
25 director directions of bar-shaped molecules and an
identical thicknesswise alignment in both regions.
The cell obtained after holding for 30 min. at 118 °C

and then cooled to room temperature, was found to retain the single-crystalline film state shown in Figure 8.

(Example 2A)

5 In order to confirm the single crystallinity of the film, an impact was applied to the cell of Example 2 after the cooling to room temperature. As a result, the crystalline film was cleaved presumably also owing to a volume shrinkage during the
10 cooling. Figure 14 is a polarizing microscope photograph (x150) showing the state. Figure 14 shows cleavage lines that extend in only three directions, and this indicates that the crystalline film had a high degree of long-distance order and was a single
15 crystal film.

(Example 3)

 The cell of Example 2A was again heated to a nematic phase temperature (130 °C) and thereafter cooled at a rate of 1 °C/min. to room temperature. In
20 the meantime, the alignment states in the nematic phase (130 °C), smectic phase (123 °C) and crystal phase (30 °C) were observed through a polarizing microscope and found to be substantially similar to those shown in Figures 4, 5 and 6, respectively.

25 (Example 4)

 The cell of Example 3 was now heated up to 118 °C at a rate of 1 °C/min. and was held at that

temperature for 30 min. similarly as in Example 2,
followed by cooling to room temperature at a rate of 1
°C/min. In the meantime, the alignment states were
observed through a polarizing microscope after the
5 holding for 1 min. and 10 min. respectively, at 118 °C
and were found to be substantially similar to those
shown in Figures 7 and 8, respectively. It was also
confirmed that the cell cooled to room temperature
retained the single-crystalline film state formed
10 after holding at 118 °C for 30 min. Thus, in the
cell, a polycrystal region as shown in a right half of
Figure 6 formed in the cell of Example 6 was
transformed into a single crystal region as shown in a
right half of Figure 8.

15 (Example 5)

A blank cell was prepared in a similar manner
as in Example 1 except for using a pair of 100 µm-
thick polymer film substrates. A sample cell was
prepared by injecting the liquid crystal material of
20 the formula (5) in a nematic phase and cooling to room
temperature in a similar manner as in Example 1.

Thereafter, the cell was again heated to 118
°C and held at the temperature for 30 min. With the
lapse of the holding time, the single crystal region
25 was observed to be enlarged until 30 min. thereafter
wherein a single-crystalline film state was enlarged
over the entire cell area of 20 mm x 20 mm. After

cooling to room temperature at a rate of 1 °C/min.,
the film in the cell exhibited a single-crystalline
film state including single crystals of 0.2 - 1 mm as
shown in a polarizing microscopic photograph (x75) of
5 Figure 15. The resultant flexible cell was applied in
a curved form along a part of the circumference of a
metal cylinder of 10 mm in diameter, and then again
subjected to the above-mentioned cycle of heating to
118 °C, holding at that temperature and cooling to
10 room temperature, whereby the curved film in the
flexible cell applied about the metal cylinder
exhibited a single-crystalline film state
substantially similar to the one shown in Figure 15.

As described above, according to the present
15 invention, an organic single-crystalline film having a
molecular alignment order provided through phase
transition from a liquid crystal phase by using an
appropriately selected liquid crystal material, and
cooling and solidifying the liquid crystal material
20 while utilizing a thickness regulating force exerted
on the liquid crystal material from a pair of
boundaries. Thus, it is possible to obtain a
functional single-crystalline film which can be
utilized in various devices.

1. A single-crystalline film having a molecular alignment order provided through phase transition from a liquid crystal phase.

2. A single-crystalline film according to Claim 1, wherein the liquid crystal phase includes a lower order liquid crystal phase and a higher order liquid crystal phase.

3. A single-crystalline film according to Claim 1, wherein the liquid crystal phase includes a smectic phase.

4. A single-crystalline film according to Claim 3, comprising a smectic liquid crystal material providing a uniform molecular alignment in a smectic layer.

5. A single-crystalline film according to Claim 4, wherein the smectic liquid crystal material has a molecular structure which is symmetrical with respect to its molecular long axis.

6. A process for producing a single-crystalline film, comprising:

a step of disposing a smectic liquid crystal

a crystallization step of cooling and solidifying the smectic liquid crystal material through its smectic phase into a single-crystalline film.

8. A process according to Claim 6, wherein the
15 crystallization step includes sub-steps of once
forming a poly-crystal state by causing phase
transition from a liquid crystal phase and
transforming the polycrystal state into a single
crystal state.

9. A process according to any one of Claims 6 - 8, wherein the crystallization step includes sub-steps of once cooling the liquid crystal material into a crystal phase and holding the liquid crystal material for a prescribed period at a temperature which is in proximity to a crystal-liquid crystal transition temperature within the crystal phase temperature range.

ABSTRACT OF THE DISCLOSURE

An organic single-crystalline film usable as a functional film in various devices is produced by selecting a liquid crystal material having a good
5 molecular alignment regularity, disposing the liquid crystal material between a pair of boundaries exerting a thickness regulating force and solidifying the liquid crystal material while imparting a molecular alignment order by phase transition from a liquid
10 crystal phase. The liquid crystal material may preferably be a smectic liquid crystal material which provides a uniform molecular alignment inclusive of the direction of molecular long axis in a smectic phase.

15

20

25

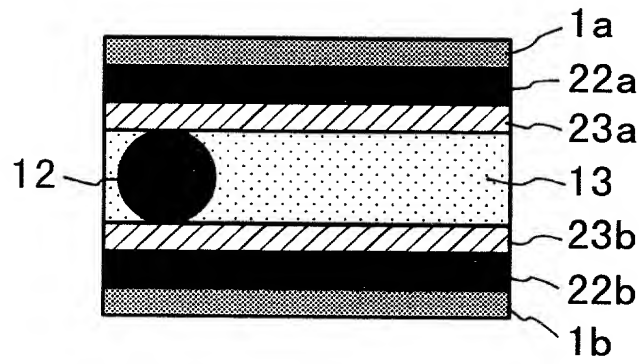


FIG. 1

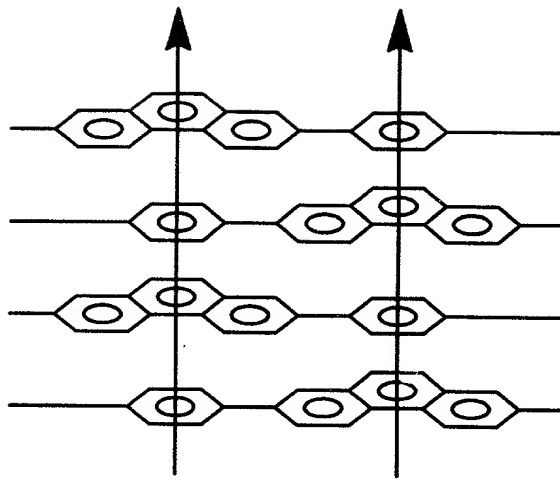


FIG. 2

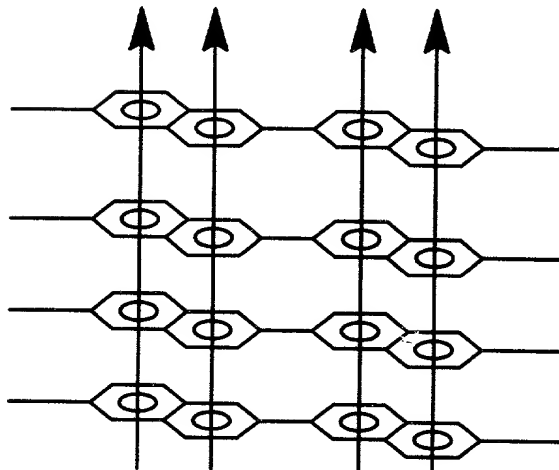


FIG. 3

A high-contrast, black and white image showing a dense, textured surface, possibly a book cover or endpaper. The texture is composed of numerous small, dark, rectangular shapes arranged in a repeating, slightly irregular pattern, creating a strong sense of depth and shadow. The overall effect is reminiscent of a woven fabric or a heavily grained material.

FIG. 5

A high-contrast, black and white photograph showing a close-up of a textured surface, possibly a book cover or endpaper. The image is characterized by a dense, grainy texture. On the left side, there is a dark, irregular shape that appears to be a shadow or a piece of tape. The right side of the image is dominated by a series of horizontal, dark, irregular shapes that look like torn paper or a series of scratches. The overall effect is one of a heavily worn and damaged surface.

A high-contrast, black and white photograph of a textured surface, possibly a book cover or endpaper. The image shows significant wear, including scratches, scuffs, and dark, irregular stains or damage. The texture is grainy and uneven, with a mix of light and dark areas. The dark areas are concentrated in the lower right and upper right, while the lighter areas are more prominent in the upper left and center. The overall appearance is one of age and wear.

FIG. 7

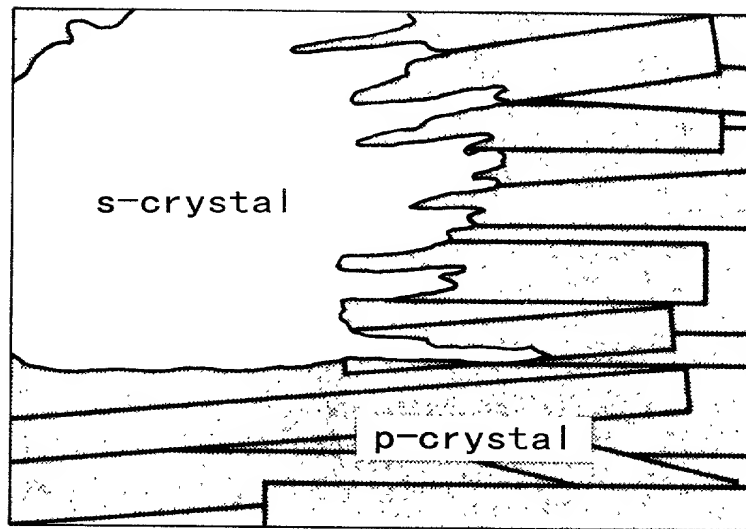
[illegible]

FIG. 9

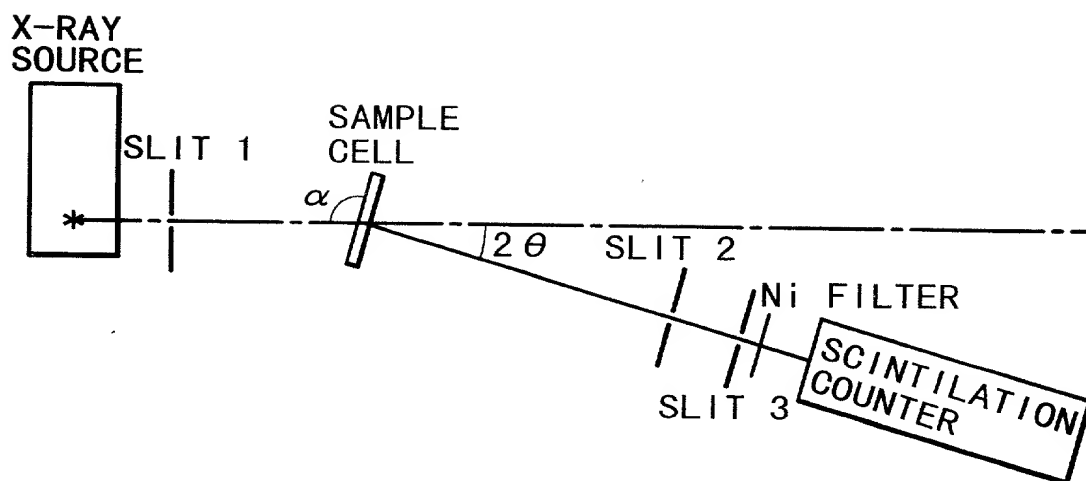


FIG. 10

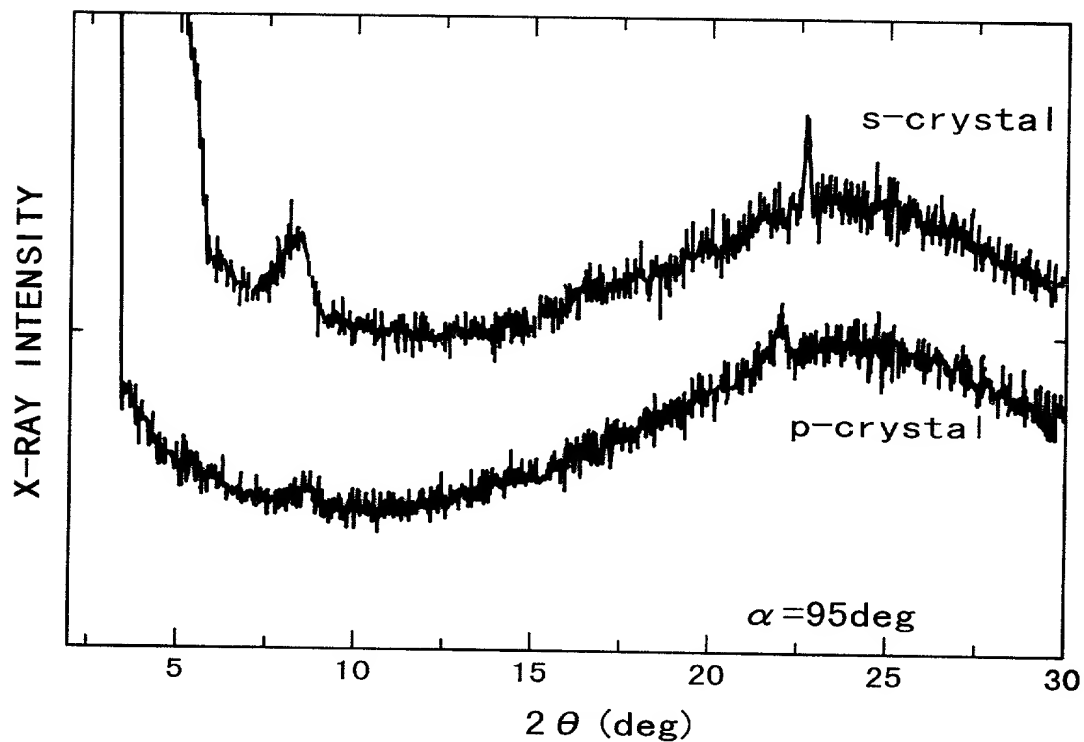


FIG. 11

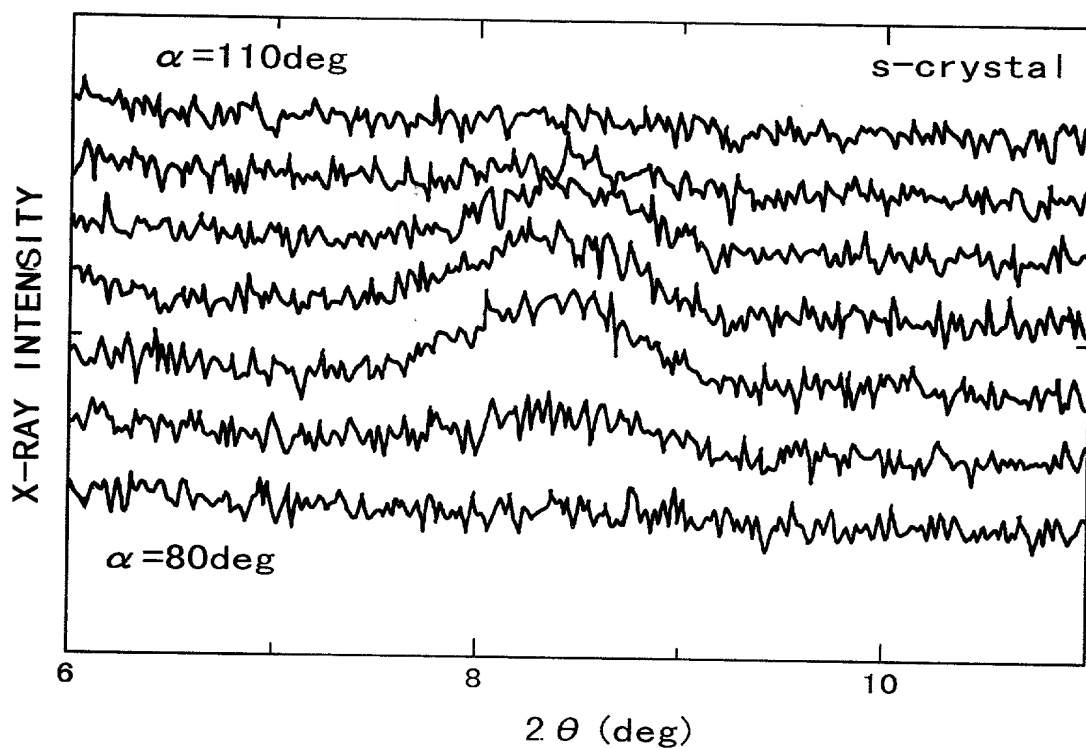


FIG. 12

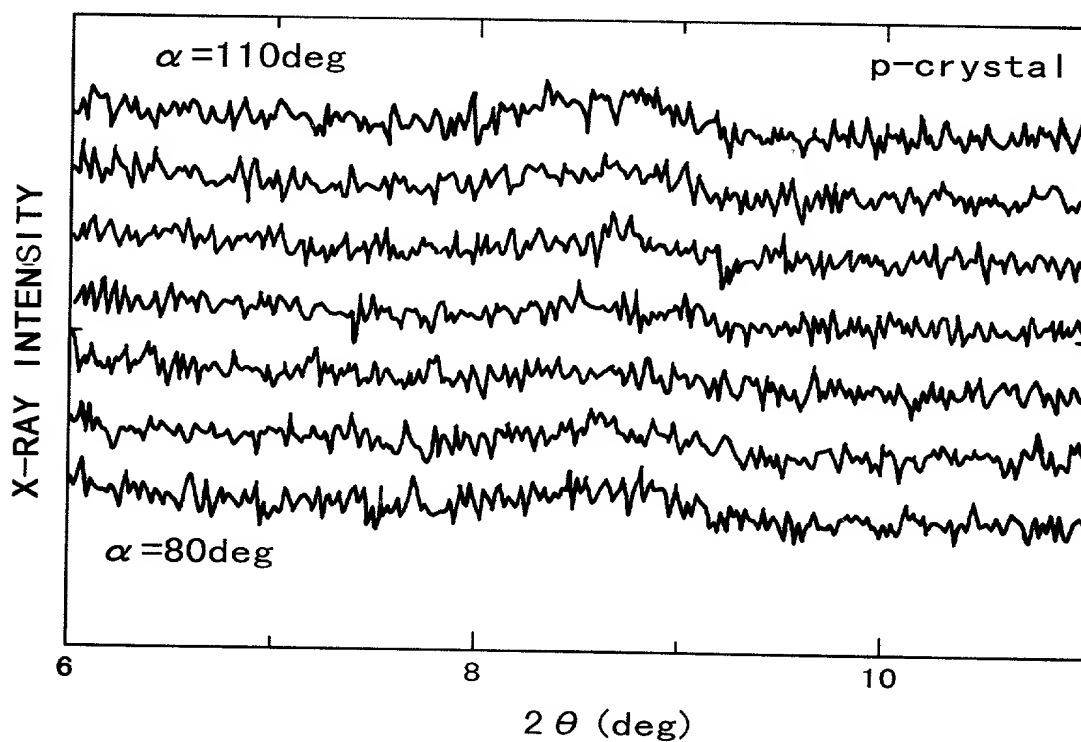


FIG. 13

A high-contrast, black and white photograph of a textured surface, possibly a wall or ceiling. The surface is covered in numerous small, dark, circular spots, likely dust or small holes. There are also larger, irregular dark shapes, possibly stains or areas of decay. The overall appearance is grainy and aged.

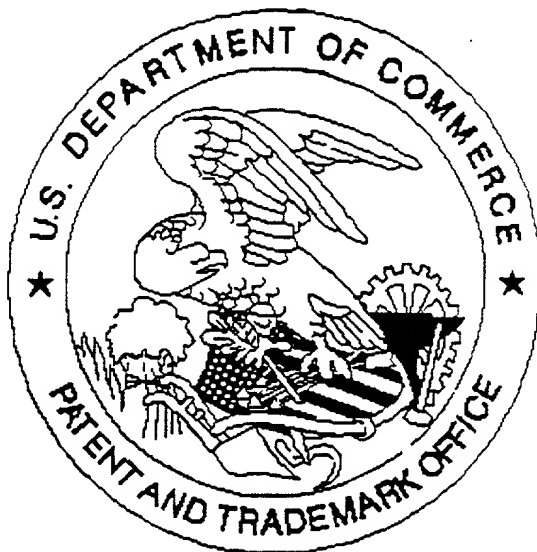
The image shows the front cover of an old book. The cover material is a dark, heavily textured cloth or buckram with a fine, woven pattern. It appears aged and worn, with visible creases, wrinkles, and some discoloration or staining, particularly along the edges and the spine area. The lighting is somewhat uneven, highlighting the texture of the material. There is no text or title visible on this side of the cover.

FIG. 14



FIG. 15

United States Patent & Trademark Office
Office of Initial Patent Examination -- Scanning Division



Application deficiencies were found during scanning:

☐ Page(s) _____ of _____ were not present
for scanning. (Document title)

☐ Page(s) _____ of _____ were not present
for scanning. (Document title)

☒ Scanned copy is best available. *Drawings.*